Quantitative Molecular Spectroscopy in Cavity

Presented by:
Technical Group Leadership:

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Technical Group Website:  
www.osa.org/EnvironmentalSensingTG

Over 1,100 Total Members

Scope:
This technical group covers optical tools and techniques used in environmental sensing, including DIAL and LIDAR, hyperspectral monitoring, detection, processing and characterization, surveying applications, atmospheric propagation, pollution monitoring, and remote imaging. Also included in this area is remote sensing for military and commercial applications such as land management, target detection, and disaster monitoring.
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- Linked-In site (global reach)
- Announce new activities
- Promote interactions
- Complement the OSA Technical Group

Member List

Activities: Webinars, Special Sessions in CLEO/FiO
Dr. Patrick Dupré, Université du Littoral Côte d'Opale

Patrick Dupré is a recognized expert in molecular high resolution and quantitative laser Spectroscopy. His career has included work in physics and chemistry laboratories in France, the United States, the United Kingdom and Germany. His interests include experimental spectroscopy and modeling. He is presently involved in developing Noise-Immune Cavity-Enhanced Optical Heterodyne Molecular Spectrometry (NICE-OHMS) for metrology applications and for trace gas detection in the Mid-InfraRed. Spectroscopy with high finesse cavity is an ideal tool for saturated absorption, i.e. under sub-Doppler conditions.
Outline

1. Motivations
2. Introduction to CEAS
3. Absorption in Cavity
4. Saturated Absorption: Modeling and Simulations
5. NICE-OHMS: Principles and Implementation
6. \( \text{C}_2\text{H}_2 \) in the NIR
7. HD in the NIR (Forbidden Transition)
8. Bibliography
9. Conclusions
Motivations

- Ultrasensitivity, i.e., Trace Detection
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- Quantitative Spectroscopy of Gas (aerosol?)
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Cavity Enhanced Absorption Spectroscopy (CEAS)

Basic Idea: Enhancing the Absorption Length, i.e., the length of interaction between light and analyte. How?
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Basic Idea: **Enhancing the Absorption Length**, i.e., the length of interaction between light and analyte. How?

- Multipass Cell
  - White Cell

Alternative:
- Resonators (using small Dichroic Mirrors, 1984)
- BBCEAS (Broad-Band Cavity-Enhanced Absorption Spectroscopy) based on Broad Band sources (coherent or not): Arc Lamps, Supercontinuum, LED, OFCS.
- It requires a Dispersive Detection
- ICOS (Integrated Cavity Output Spectroscopy), On-Axis, vs. Off-Axis
- CRDS (Cavity Ring-Down Spectroscopy), Continuous or Pulsed Wave, Broad-Band vs. Narrow-Band Source (see O'Keefe 1988)
- Cavity Finesse measurement (in Frequency)
- Cavity Impedance Mismatch (Ring)
- FMS (Frequency Modulation Spectroscopy)
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Off-Axis ICOS with recycling mirror to recover the leaking input power
(with the permission of J. Mandon)
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With the permission of C. Vallance
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Absorption

\[ I_0 I(\omega) \]

Source

Detector

\[ L_{abs} \]
The Absorption Beer-Lambert Law:

\[ I(\omega) = I_0 e^{-\alpha(\omega) L_{\text{abs}}} \]

\( \alpha(\omega) = N \sigma(\omega) \) is the frequency depending absorption coef. (in cm\(^{-1}\)).
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Approximation of the Optically Thin Medium [\( \alpha(\omega) L_{\text{abs}} \ll 1 \)]:

\[ \Delta I(\omega) = \frac{I_0 - I(\omega)}{I_0} = \alpha(\omega) L_{\text{abs}} \]
Background

- The Absorption Beer-Lambert Law:
  \[ I(\omega) = I_0 e^{-\alpha(\omega)L_{abs}} \]

  \[ \alpha(\omega) = \mathcal{N} \sigma(\omega) \] is the frequency depending absorption coefficient (in cm\(^{-1}\)).

- Approximation of the Optically Thin Medium \([\alpha(\omega)L_{abs} \ll 1]\):
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The Absorption Beer-Lambert Law:

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- the Gas Pressure
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The Number Density \( (N \text{ in cm}^{-3}) \) is proportional to

- the Gas Pressure
- the Concentration of each specific species
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The Number Density \((N \text{ in cm}^{-3})\) is proportional to

- the Gas Pressure
- the Concentration of each specific species

The line absorption Cross-section \((\sigma(\omega) \text{ in cm}^2/\text{molecule})\), includes a Normalized Lineshape (like a Voigt profile whose width is pressure depending):

\[ \int \sigma(\omega) \, d\omega = S \]

where \(S\) is the Line Intensity (in cm/molecule if \(\omega\) is in cm\(^{-1}\)).
The Absorption Beer-Lambert Law:

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\(S\) is available in the database like HITRAN \([\text{http://hitran.org/}]\)
Absorption in a Resonant Cavity

\[ I_0(\omega) \]

\[ L_{abs} \]

Source

ADC

Computer

Detector

ADC

Computer

Spectroscopy in Cavity Dec. 2018 10 / 47
Main Features of a Symmetric Resonant Cavity

- Cavity Finesse (Enhancement Factor): $F = \frac{\pi}{L}$
- Free Spectral Range (FSR): $FSR = \frac{c}{L_{cav}}$
- Response Time (or Characteristics Time): $\tau_{RD} = F \frac{L_{cav}}{\pi c} = \frac{F}{2} \frac{\pi}{t_{rt}} = \frac{1}{2} \frac{\Delta c_{cav}}{FSR}$
- Equivalent Absorption Length: $L_{eq} = \frac{2}{F} \frac{\pi}{L_{cav}} = \frac{2}{c \tau_{RD}}$
- Trapped Power: $I_{cav} = = \frac{F}{\pi} I_{in}$
Main Features of a Symmetric Resonant Cavity

- Cavity Finesse (Enhancement Factor)

\[ F = \frac{2\pi}{L} = \frac{\pi \sqrt{R}}{1 - R} \]
Main Features of a Symmetric Resonant Cavity

- Cavity Finesse (Enhancement Factor)
  \[ F = \frac{2\pi}{\mathcal{L}} = \frac{\pi \sqrt{R}}{1 - R} \]

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  \[ FSR = \frac{c}{2L_{cav}} \]
Main Features of a Symmetric Resonant Cavity

- **Cavity Finesse (Enhancement Factor)**

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\]

- **Free Spectral Range (FSR)**

\[
FSR = \frac{c}{2L_{cav}}
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- **Response Time (or Characteristics Time)**

\[
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- Equivalent Absorption Length
  \[ L_{eq} = \frac{2\mathcal{F} L_{cav}}{\pi} = 2c \tau_{RD} \]

- Trapped Power
  \[ I_{cav} = \frac{\mathcal{F} I_{in}}{\pi} \]
Formalism: Transfer Function (“Filter”) of a Lossless Cavity

In the Frequency Domain

$I_{\text{out}}(\omega) = |T_{\text{cav}}(\omega) \cdot E_{\text{in}}(\omega)|^2$

with (obtained from multiple interferences)

$T_{\text{cav}}(\omega) = T_{e} - i\omega t_{rt}/2 + R \sum_{i}^{1} (\omega - i\omega_{\text{FSR}})\Delta_{\text{cav}}(\omega)$

and with

$R' = R_{e} - \alpha(\omega)L_{\text{abs}}$

In the Time Domain

$I_{\text{out}}(t) = \left| FT^{-1}[T_{\text{cav}}(\omega) \cdot E_{\text{in}}(\omega)] \right|^2$
Formalism: Transfer Function ("Filter") of a Lossless Cavity

- In the Frequency Domain

\[ I_{out}(\omega) = |T_{cav}(\omega) \cdot E_{in}(\omega)|^2 \]

with (obtained from multiple interferences)

\[ T_{cav}(\omega) = \frac{T \, e^{-i\omega t_r / 2}}{1 + R \, e^{i\omega t_r}} = \frac{T}{1 - R} \sum_i \frac{1}{1 + i \left( \frac{\omega - i \omega_{FSR}}{\Delta_{cav}(\omega)} \right)} \]

and with

\[ R' = R \, e^{-\alpha(\omega) L_{abs}} \]
Formalism: Transfer Function (“Filter”) of a Lossless Cavity

- **In the Frequency Domain**

\[ I_{out}(\omega) = |T_{cav}(\omega) \cdot E_{in}(\omega)|^2 \]

with (obtained from multiple interferences)

\[ T_{cav}(\omega) = \frac{T e^{-i\omega trt/2}}{1 + Re^{i\omega trt}} = \frac{T}{1 - R} \sum_i \frac{1}{1 + i\left(\frac{\omega - i\omega_{FSR}}{\Delta_{cav}(\omega)}\right)} \]

and with

\[ R' = Re^{-\alpha(\omega)L_{abs}} \]

- **In the Time Domain**

\[ I_{out}(t) = \left| FT^{-1}\left[T_{cav}(\omega) \cdot E_{in}(\omega)\right]\right|^2 \]
Simulation: Pulsed Source

Case of a Fourier Transform Limited Pulsed Laser

Frequency Space

|E_{in}|^2

Δω_L

ω_L

ω

|F|^2

Δω

FSR

ω_{0p}

ω

|E_{out}|^2

Δω_L > FSR

ω_L

ω

Time Space

|E_{in}|^2

t_{coh}

INPUT

|f|^2

τ_e

t_{rt}

FILTER

|E_{out}|^2

t_{coh} < t_{rt} < τ_e

OUTPUT
Simulation: CW Source

Case of a CW Laser

\[ \Delta \omega < \Delta \omega_L < \text{FSR} \]

Frequency Space

\[ |E_{in}|^2 \quad \Delta \omega_L \]

\[ F \quad \text{FSR} \quad \Delta \omega \]

Time Space

\[ |E_{in}|^2 \quad t_{coh} \]

\[ |f|^2 \quad t_{rt} \quad \tau_e \]

\[ |E_{out}|^2 \quad t_{rt} < t_{coh}, t_{sw} \ll \tau_e \]

INPUT

FILTER

OUTPUT
Absorption in a Resonant Cavity

Linear Absorption at resonance (occupancy factor $\mu = 1$)

$$\tau_{RD}(\omega) - \tau_0 = \alpha(\omega)c$$

Alteration of the Detected Power

$$\Delta I(\omega) = \alpha(\omega)L_{eq}$$

NonLinear Absorption

$$\alpha(\omega, I)$$

Alteration of the Decay Shape (to nonexponential decay)

Lamb-dip, etc...

P. DUPRÉ (pdupre@gmx.com) (LPCA/ULCO)

Spectroscopy in Cavity

Dec. 2018
Absorption in a Resonant Cavity

- Linear Absorption at resonance (occupancy factor = 1)
Absorption in a Resonant Cavity

- **Linear Absorption** at resonance (occupancy factor = 1)
  - Alteration of the Characteristics Time (CRDS)

\[
\frac{1}{\tau_{RD}(\omega)} - \frac{1}{\tau_0} = \alpha(\omega) \ c
\]
Absorption in a Resonant Cavity

- **Linear Absorption** at resonance (occupancy factor = 1)
  - Alteration of the Characteristics Time (CRDS)

\[
\frac{1}{\tau_{RD}(\omega)} - \frac{1}{\tau_0} = \alpha(\omega) \ c
\]

- Alteration of the Detected Power

\[
\frac{\Delta I(\omega)}{I_0} = \alpha(\omega) \ L_{eq}
\]
**Linear Absorption** at resonance (occupancy factor = 1)
- Alteration of the Characteristics Time (CRDS)

\[
\frac{1}{\tau_{RD}(\omega)} - \frac{1}{\tau_0} = \alpha(\omega) \cdot c
\]

- Alteration of the Detected Power

\[
\frac{\Delta I(\omega)}{I_0} = \alpha(\omega) \cdot L_{eq}
\]

**NonLinear Absorption**
Absorption in a Resonant Cavity

- **Linear Absorption** at resonance (occupancy factor = 1)
  - Alteration of the Characteristics Time (CRDS)
    \[
    \frac{1}{\tau_{RD}(\omega)} - \frac{1}{\tau_0} = \alpha(\omega) c
    \]
  - Alteration of the Detected Power
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- **NonLinear Absorption**
  - \(\alpha(\omega, I)\)
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Limit of Detection or Sensitivity

Analysis of the Signal to Noise Ratios (SNR) for the different techniques

\[
\text{Signal (Cavity Enhancement factor: } \sim F \text{)}
\]

\[
\text{Source Intensity Fluctuations versus Photon-Shot-Noise (PSN)}
\]

\[
\frac{\Delta \nu}{\eta P_{\text{eff}}} = \left( \alpha L_{\text{eq}} \right)_{\text{min}}
\]

CRDS is intrinsically Immune to Source Intensity Fluctuations (discontinuous acquisition)

The “Direct” Absorption techniques require acquiring the Noise Immunity

Differential Absorption (DAS)
Amplitude Modulation
Frequency/Phase Modulation (FMS)
Beam Intensity Stabilization (AOM)

NICE-OHMS benefits of both: CW acquisition, and full noise Immunity.
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- **Signal** (Cavity Enhancement factor: $\sim F$)
- **Source Intensity Fluctuations versus Photon-Shot-Noise (PSN)**

$$\sqrt{\frac{2 e \Delta \nu}{\eta \langle P_{eff} \rangle}} = (\alpha L_{eq})_{min}$$
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S. Hu (Hefei) in 2017 (CO), Rev. Scient. Inst., 88, 043108

Applications:

High Resolution Spectroscopy

Simultaneous determination of the number density and of the cross section, from a single decay (CRDS)!

Attention

Requiring full control of the Intracavity Power

Modeling of the Nonlinear Interaction

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P. DUPRÉ (pdupre@gmx.com) (LPCA/ULCO)
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  - Crossover Resonances
NonExponential Decay

CRDS of Jet-Cooled NO₂: Decay of the line at 12536.4464 cm\(^{-1}\) (\(^9\)R\(_0\)(0), 3/2)

\[ \tau_{rd} = 136.1 \mu s \]
\[ \tau_{rd} = 129.2 \mu s \]
\[ \tau_{rd} = 121.8 \mu s \]
\[ \tau_{rd} = 115.0 \mu s \]
\[ \tau_e = 152.7 (5) \mu s \]

ROC: 0.5 m, \(L = 0.35\) m, \(w_0 = 0.246\) mm
\(T = 4.5\) ppm, \(R_F = 10\) k\(\Omega\), \(\eta = 0.55\) A/W

\[ \tau_{rd} = 106.7 \mu s \]
\[ \tau_{rd0} = 106.0 (1.5) \mu s \]

UnWeighted: \(\tau_0 = 275 (4) \mu s, I_{sat} = 38.8 (2.8)\) MW/m\(^2\)

Weighted: \(\tau_0 = 347 (2) \mu s, I_{sat} = 99.5 (2.5)\) MW/m\(^2\)
Saturation in NO₂ (with Fine and Hyperfine Transitions)

Power dependence of the $^4Q_{21}(0.5)$ Line Pattern

Excitation Energy (cm$^{-1}$)

Absorption (/cm)

Linear\textsuperscript{a}

1.5 W\textsuperscript{a}

3 W\textsuperscript{b}

3.7 W\textsuperscript{a}

9.2 W\textsuperscript{a}

19 W\textsuperscript{b}

46 W\textsuperscript{b}

92 W\textsuperscript{c}

230 W\textsuperscript{c}

Voigt

0.5 W

1.5 W

3 W

6 W

10 W

20 W

50 W

100 W

250 W

Excitation Energy (cm$^{-1}$)
Saturation in NO$_2$ (with Fine and Hyperfine Transitions)

**Power dependence of the $^q_0Q_{21}(0.5)$ Line Pattern**

- Linear
  - 0.6 W
  - 1.5 W
  - 3 W
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  - 9.2 W
  - 19 W
  - 46 W
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  - 230 W

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  - 0.5 W
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  - 3 W
  - 6 W
  - 10 W
  - 20 W
  - 50 W
  - 100 W
  - 250 W

**Power dependence of the $^q_0R_{11}(0.5)$ Line Pattern**

- Linear
  - 0.6 W
  - 1.5 W
  - 3 W
  - 3.7 W
  - 6.0 W
  - 7.5 W
  - 9.2 W
  - 15 W
  - 19 W
  - 37 W
  - 46 W
  - 92 W
  - 230 W

- Voigt
  - 0.5 W
  - 1.5 W
  - 3 W
  - 6 W
  - 10 W
  - 20 W
  - 50 W
  - 100 W
  - 250 W
Absorption versus the Intracavity Power at the Center of the $^qR_{11}(0.5)$ Line Pattern

- Experiment
- Uniform Weighting
- STD Weighting

<table>
<thead>
<tr>
<th>Running Wave Power (W)</th>
<th>Absorption (/cm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1×10^{-6}</td>
<td></td>
</tr>
<tr>
<td>2×10^{-6}</td>
<td></td>
</tr>
<tr>
<td>3×10^{-6}</td>
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<tr>
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Absorption versus Intracavity Power
NICE-OHMS: History in a nutshell

Pioneerly developed at NIST (J. Hall, J. Ye), first publication in 1996 on acetylene at 1.064 \( \mu \)m (Nd:YAG/Ti:Sa), cavity finesse: \( \sim 100000 \), \( \text{NEA} \sim 1 \times 10^{-14} \text{cm}^{-1}/\text{Hz} \).

Since 2007: Ove Axner group (Umeå, SW), all fibered NIR (EDFL and DFB), MIR (OPO), more than 18 Publications (\( F \sim 50000 \)), \( \text{NEA} \sim 4 \times 10^{-13} \text{cm}^{-1}/\text{Hz} \).

Technical Developments;
2017: Whispering-Gallery-Mode Laser

Since 2010: Ben McCall (UIUC, IL), Molecular Ion (Spectroscopy), Ti:Sa (Red), DFG and OPO (MIR), Jet Expansion, cavities of modest Finesse

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Since 2015: National Tsing Hua University (Taiwan), Quantum-Dot ECDL at 1.283 µm ($\text{N}_2\text{O}$ in Doppler), + CRDS, Atomic ParityNonConservation

Since 2016: Collaboration Dunkerque/Amsterdam (VU), Metrology of Hydrogen
NICE-OHMS: History in a nutshell

- **Pioneerly developed at NIST** (J. Hall, J. Ye), first publication in 1996 on acetylene at 1.064 μm (Nd:YAG/Ti:Sa), cavity finesse: \( \sim 100000 \), NEA \( \sim 1 \times 10^{-14} \text{ cm}^{-1} / \sqrt{\text{Hz}} \)

- Since 2007: Ove Axner group (Umeå, SW), all fibered NIR (EDFL and DFB), MIR (OPO), more than 18 Publications \( (\mathcal{F} \sim 50000) \), NEA \( \sim 4 \times 10^{-13} \text{ cm}^{-1} / \sqrt{\text{Hz}} \), **Technical Developments**: 2017: Whispering-Gallery-Mode Laser

- Since 2010: Ben McCall (UIUC, IL), **Molecular Ion** (Spectroscopy), Ti:Sa (Red), DFG and OPO (MIR), Jet Expansion, cavities of modest Finesse

- Since 2013, Frans Harren (Radboud Univ., Nijmegen, NL), NIR, **Trace Detection**

- Since 2014, Livio Gianfrani (Naples Univ.): ECDL at 1.39 μm \( (\text{H}_2^{18}\text{O}) \), **Metrology**: Boltzmann Constant, Symmetrization postulate (detec. of forbidden transition), HD?

- Since 2015: Dual NICE-OHMS (CO, NIR), Shally Saraf, Robert Byer (Stanford University, CA), **Metrology** (Testing Lorentz Invariance, STAR Project)?

- Since 2015: National Tsing Hua University (Taiwan), Quantum-Dot ECDL at 1.283 μm \( (\text{N}_2\text{O in Doppler}) \), + CRDS, **Atomic ParityNonConservation**

- Since 2016: Collaboration Dunkerque/Amsterdam (VU), **Metrology** of Hydrogen

- Since 2017: Stefan Schäffer, Niels Bohr Institute (Copenhagen), **MOT** of \(^{88}\text{Sr}\) (locking against transition)
Saturation Modeling: an Insight

- Establishing the complex absorption in the Frequency domain (coupling of the EMF with the susceptibility) for a given Doppler Shift
Saturation Modeling: an Insight

- Establishing the complex absorption in the Frequency domain (coupling of the EMF with the susceptibility) for a given Doppler Shift
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- Establishing the Linear Absorption

Gaussian (Impact Parameter)
Transit-Time Broadening (vs. Power Broadening)
Approximation based on the spectral extension of products involved (⊗, ×)
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- Establishing the Saturated Absorption (based on the Rabi Frequency)
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Establishing the Linear Absorption

Establishing the Saturated Absorption (based on the Rabi Frequency)

Plugging specific EMFs (FMS) and considering the Stationary Response
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- Establishing the Linear Absorption
- Establishing the Saturated Absorption (based on the Rabi Frequency)
- Plugging specific EMFs (FMS) and considering the Stationary Response
  - Monochromatic (Radial Extension)

Numerical Integration over the Doppler Shift
Integration over the Impact Parameter
Integration over the Transit-Time Rate (Maxwell Boltzmann)
Summation of the Degenerated Zeeman Sub-Transitions (Polarization)
No Saturation Coefficient is used
Saturation Modeling: an Insight

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- No Saturation Coefficient is used
Simulation and Line Profile Analysis

**Saturation Analysis:** $\text{C}_2\text{H}_2$ Transition $R(0)$ at 6558.79233 cm$^{-1}$ (polyad 10)

- **Pressure:** ~ 1.7 Pa
- **Transit-time Rate:** 160 kHz
- **Collisional Rate:** 100 kHz
- **Dipole Moment:** ~ 11 mD
- **Beam Waist:** 0.45 mm
- **Beam Power:** 10 mW
- **Doppler Broadening:** 0.00793 cm$^{-1}$

![Graph showing absorption spectra with different integration methods and parameters.](image-url)
C$_2$H$_2$ NICE-OHMS Simulation (Absorption)

*NICE-OHMS in Phase, C$_2$H$_2$ Transition R(0) at 7143.8289 cm$^{-1}$ (polyad 11)*

- Dipole Moment: ~ 0.912 mD
- Beam Waist: 0.46 mm
- Beam Power: 200 mW
- Modulation Frequency: 350 MHz
- Modulation Index: 0.4
- Rabi Frequency: 97.8 kHz
- Pressure: ~ 1 Pa
- Transit-time Rate: 220 kHz
- Collisional Rate: 60 kHz
- Doppler Broadening: 0.00863 cm$^{-1}$
C$_2$H$_2$ NICE-OHMS Simulation (Dispersion)

*NICE-OHMS in Quadrature, C$_2$H$_2$ Transition R(0) at 7143.8289 cm$^{-1}$ (polyad 11)*

- **Dipole Moment:** $\sim 0.912$ mD
- **Beam Waist:** 0.46 mm
- **Beam Power:** 200 mW
- **Modulation Frequency:** 350 MHz
- **Modulation Index:** 0.4
- **Mean Rabi Frequency:** 97.8 kHz

- **Pressure:** $\sim 1$ Pa
- **Mean Transit-time Rate:** 220 kHz
- **Collisional Rate:** 60 kHz
- **Doppler Broadening:** 0.00863 cm$^{-1}$

![Graph showing dispersion vs. wavelength with key parameters indicated.]
**C₂H₂ Simulation (Dispersion): Lorentzian Component**

*Saturation Analysis: C₂H₂ Transition R(0) at 7143.8289 cm⁻¹ (polyad 11)*

- **Dipole Moment:** 0.912 mD
- **Power:** 200 mW, **Waist:** 0.46 mm
- **Linear Polarization**
- **Modulation Index:** 0.4

**Graph:**
- **Lorentzian Component Width (MHz)** vs **Collision Rate (kHz)**
- **Transit-time Rate:** 220 kHz
- **Rabi Frequency:** 97.8 kHz
- **Double Integration**
- **Triple Integration**
Saturation Analysis: \( \text{C}_2\text{H}_2 \) Transition \( R(0) \) at 7143.8289 cm\(^{-1}\) (polyad 11)

- Dipole Moment: 0.912 mD
- Power: 200 mW, Waist: 0.46 mm
- Linear Polarization
- Modulation Index: 0.4

Gaussian Component Width (MHz) vs. Collision Rate (kHz)

- Double Integration
- Triple Integration

Rabi Frequency: 97.8 kHz
Transit-time Rate: 220 kHz
Phase Modulations in NICE–OHMS
Phase Modulations in NICE–OHMS
Frequency Modulation in Cavity

Phase Modulations in NICE–OHMS
Phase Modulations in NICE–OHMS
NICE-OHMS Implementation
NICE-OHMS Implementation

Laser source
ECDL

Immersed Cavity
NICE-OHMS Implementation

Laser source (ECDL) → EOM → Immersed Cavity

- Frequency Error to Voltage Converter
- PZT
- PDH
- FSR
- EOM
- PID

Frequency Error

~20 MHz
~310 MHz

Immersed Cavity

PZT

Frequency Error to Voltage Converter
NICE-OHMS Implementation

Laser source
ECDL

EOM

Immersed Cavity
PZT

ADC
Computer

f_{FSR} \sim 310 \text{ MHz}

f_{PDH} \sim 20\text{ MHz}
NICE-OHMS Implementation

Immersed Cavity

Frequency Error to Voltage Converter

PZT

Laser source (ECDL)

ADC

Computer

Lockin Amplifier

f_{mod.}

f_{FSR} \sim 310 \text{ MHz}

f_{PDH} \sim 20 \text{ MHz}

EOM

Immersed Cavity

PID

I
NICE-OHMS Implementation

- Laser source: ECDL
- Optical Frequency Comb
- Cs clock, 10^{-13} accuracy
- Beatnote unit
- Counter
- Frequency Error to Voltage Converter
- Lockin Amplifier
- ADC Computer
- Immerged Cavity

Key frequencies:
- \( f_{\text{PDH}} \approx 20 \text{MHz} \)
- \( f_{\text{FSR}} \approx 310 \text{ MHz} \)
- \( f_{\text{PDH}} \approx 20 \text{MHz} \)
- \( f_{\text{ref}} \)
- \( f_{\text{beat}} \)
- \( f_{\text{mod.}} \)
Direct Absorption of C$_2$H$_2$ (P 11, $\nu_1 + \nu_2 + (2\nu_4 + \nu_5)^1 \leftarrow 0$)

C$_2$H$_2$, Transition $R_e(4)$ (7239.79077 cm$^{-1}$), Direct Absorption at 7 µbar

Intra-Cavity Power (W)

Transit-Time Rate: 222 kHz
Dip. Moment.: 0.482 mD

$L_{eq}$: ~27.2 km
Direct Absorption of C$_2$H$_2$ (P 11, $\nu_1 + \nu_2 + (2\nu_4 + \nu_5)^1 \leftarrow 0$)

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Doppler: 519.7 (6) MHz  
HWHM: 363 (71) kHz  
Doppler: 519.7 (6) MHz
Direct Absorption of C$_2$H$_2$ (P 11, $\nu_1 + \nu_2 + (2\nu_4 + \nu_5)^1 \leftarrow 0$)

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HWHM: 363 (71) kHz

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Absorbance
Intra-Cavity Power (W)

Detuning (MHz)
NICE-OHMS “Absorption” of C$_2$H$_2$ (Polyad 11)

C$_2$H$_2$, Transition $R_e(4)$ (7239.79077 cm$^{-1}$), NICE-OHMS in Phase at 7 µbar

Intra-Cavity Power: ~39 W
Freq. Mod.: 311.741 MHz (Mod. Index: 0.3)
Effective $\mu_{band}$: 0.482 mD
Transit-Time Rate: 222 kHz

$\Gamma_L = 341 (4)$ kHz
NICE-OHMS “Dispersion” of C$_2$H$_2$ (Polyad 11)

C$_2$H$_2$, Transition $R_e(4)$ (7239.79077 cm$^{-1}$), NICE-OHMS in Quadrature at 7 µbar

Intra-Cavity Power: ~39 W
FSR: 311.741 MHz (Mod. Index: 0.3)
Effective $\mu_{band}$: 0.482 mD
Transit-Time Rate: 222 kHz

$\Gamma_L = 393 (4)$ kHz
NICE-OHMS Dispersion of C$_2$H$_2$ (Polyad 11) with OFC

C$_2$H$_2$, Transition $R_e$(4) (7239.79077 cm$^{-1}$), NICE-OHMS in Quadrature at 2 $\mu$bar

No Dithering

Dithering 1f (596.5 Hz?)

Dithering 2f

$\nu_0 = 217043458142$ (15) kHz
$\Gamma = 314.4$ (12.4) kHz

$\nu_0 = 217043458145$ (3.8) kHz
$\Gamma = 318.7$ (3.9) kHz

$\nu_0 = 217043458130$ (4.9) kHz
$\Gamma = 316.8$ (4.9) kHz
Resonance Width Power Dependence

Power Broadening, NICE-OHMS of $\text{C}_2\text{H}_2$, Transition $R_2$, $\nu_1 + \nu_2 + (2\nu_4 + \nu_5)^1$
Outline

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2. Introduction to CEAS
3. Absorption in Cavity
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6. C$_2$H$_2$ in the NIR
7. HD in the NIR (Forbidden Transition)
8. Bibliography
9. Conclusions
Simulation of HD: First Overtone, Transition $R(0)$

- Excitation Energy (cm$^{-1}$)
- "Dispersion" for 1 cm
- Linear
- 2 MHz

Graph showing the dispersion for 1 cm with excitation energy in cm$^{-1}$.
1-f WM-NICE-OHMS: HD Transition $R_1$ of the 1st Overtone (1 Pa)

Intracavity Power: ~ 138 W
Cavity Finesse: ~ 125000
$L_{eq} \sim 75$ km
HD Ground State: Hyperfine Energy Levels for N~1

According to Ramsey and Lewis
Discussion on HD (Work in Progress)


Improvement by 3 Orders of Magnitude

R₁, R₂, R₃ observed

Sensitivity ∼ 10^{-12} cm⁻¹

Resonance Line Shape (Asymmetry)?

Mean Transit-Time Rate: ∼ 660 kHz

Rabi Frequency (∼ 21 kHz)?

Recoil (∼ 35 kHz)?

Evidence of the Hyperfine Structure (spreads over 600 kHz)?

Pressure Broadening Coefficient?

Pressure Shift?

Comparison with CRDS (S. Hu group, in PRL)
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- Improvement by 3 Orders of Magnitude

Improvement by 3 Orders of Magnitude

$R(1), R(2), R(3)$ observed
Discussion on HD (Work in Progress)

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(published value: 217.105 192 (30) THz [Kassi/Campargue JMS, 2011])

Improvement by 3 Orders of Magnitude

\(R(1), R(2), R(3)\) observed

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Resonance Line Shape (Asymmetry)?
- Mean Transit-Time Rate: \(\sim 660 \text{ kHz}\)
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Bibliography

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- HD: $R(0)$, $P(1)$, etc.

Higher Finesse: from $\sim 125000$ to $500000$?

Full Validation of the NICE-OHMS Technique on $C_2H_2$

Saturation Model Validation?

Line Shape asymmetries: Role of the Hyperfine Structure?

Resonance Narrowing?

Sub-kHz Precision Range?

Etc.
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Thank for your Attention